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# Electro-optical properties of aligned polymer dispersed liquid crystal films

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## Abstract

We study the electro-optical properties of polymer dispersed liquid crystal (PDLC) films with various kinds of alignment. These films are transparent in the electric field-on state and opaque in the field-off state. Experimental data show that the contrast ratio of these aligned films can be higher than that of a usual type PDLC film. The relationship between the field-off state transmittance of such a PDLC film and the polarization of the incident light is discussed. The optical properties of such aligned films can be described by a theory based on Beer's law. © 1997 Published by Elsevier Science B.V.

## 1. Introduction

Polymer dispersed liquid crystal (PDLC) film, which consists of micron-sized nematic liquid crystal (NLC) droplets dispersed in a polymer matrix, shows an interesting optical property based on voltage-controlled light scattering by nematic droplets. Normal-mode operation of PDLC film is characterized by two optically different states, an opaque state under no applied voltage (the off-state) and a transparent state under applied voltage (the on-state) [1–6]. The off-state transmittance of a PDLC usually has maximum refractive index mismatching such that the scattering is at a maximum and the transmittance is at a minimum. Contrary to the off-state, in the case of on-state, the transmittance is at a maximum because of the refractive index matching. The ratio of the minimum and maximum transmittances is called the contrast ratio. Comparing to a commercial NLC

film, the contrast ratio of current PDLC films is not high enough. Much effort is still needed for improving the contrast ratio of a PDLC film.

Recently, it has been proposed that a PDLC film with dye dissolved in NLC possesses a controllable absorbance as well as a controllable scattering [7–10]. This combination is proved to have some merit for improving the contrast ratio, and is reported in our previous report [10]. In addition, we report here that a conventional PDLC can be aligned by rubbing the substrate of the film. The alignment for the PDLC film can also improve the contrast ratio.

We fabricated the PDLC films by using a UV curable polymer as a PDLC material, and to cure the film at suitable UV intensity [10]. Three kinds of films, plane-parallel, twisted and not aligned PDLC films, were made. In the on-state, the symmetry axis of every NLC droplet of these three kinds of PDLC films, as shown in Fig. 1a, is normal to the sub-

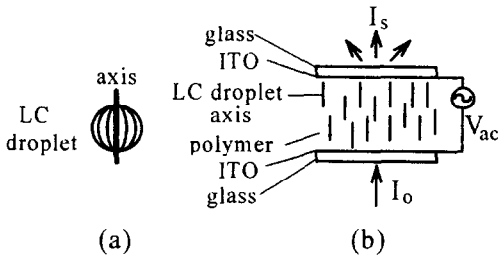


Fig. 1. (a) A liquid crystal droplet. The nematic liquid crystal molecules are aligned along the line drawn, and the alignment is symmetric with respect to the droplet axis. (b) In the on-state, most of  $I_0$  transmits through the PDLC film.

strates. Because the refractive indices of NLC and polymer match each other, the NLC droplets of these three kinds of PDLC films scatter little incident light. Therefore, most of the incident light can pass through the PDLC films. However, in the off-state, most of the droplet axes are not normal to the substrate. Because of the refractive index mismatch between the NLC and polymer, the NLC droplets scatter the incident light.

Comparing the electro-optical properties of these PDLC films, we find that the on-state transmittances of these three kinds of films are almost the same, but the off-state transmittances of these three kinds of films are different from each other. The off-state transmittance of an aligned PDLC film is dependent on the polarization direction of the incident light. We evaluate the contrast ratio of each kind of aligned films and find that for normal incident natural light, the twist aligned PDLC film is better than a conventional PDLC film which has no alignment. However, for a linearly polarized incident light, the parallel alignment can have the best contrast ratio. The electro-optical properties of such aligned films are described by a theory based on Beer's law. Details of the experimental results and theoretical explanation are reported in this paper.

## 2. Theory

We consider a PDLC film as shown in Fig. 1b. There are many nematic liquid crystal (NLC) droplets which are randomly distributed in the film. The ordinary and extraordinary refractive indices of the NLC are  $n_o$  and  $n_e$ , respectively. Outside these NLC

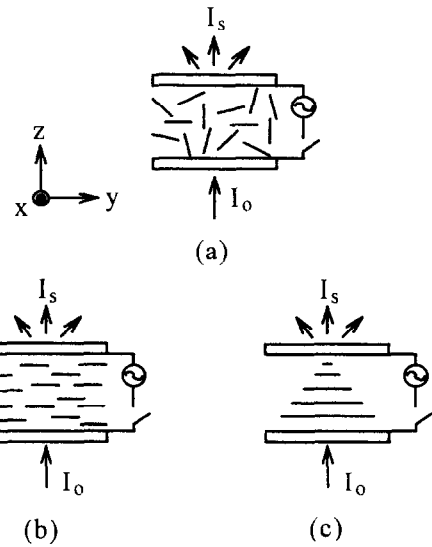


Fig. 2. In the off-state, most of the NLC droplets' axes are not normal to the glass substrates. The incident light can be scattered by these PDLC films. (a) A conventional PDLC film, (b) a plane-parallel aligned PDLC film, (c) a twist aligned PDLC film.

droplets is polymer in solid phase with refractive index  $n_p \approx n_o$ . In the on-state, the symmetry axes of NLC droplets are directed in only one direction, and because  $n_p \approx n_o$  most of the incident light  $I_0$  transmits through the PDLC film. In the off-state, as shown in Fig. 2, most of the axes are not normal to the glass substrates. And because of the refractive index mismatching of the NLC and the polymer, the incident light  $I_0$  is scattered by the NLC droplets. The transmittance  $T$  of normal incident light can be measured by using the setup shown in Fig. 3. The visible light source can be a He-Ne laser. Because polymer and NLC molecules absorb nearly no visible light, the absorption of the PDLC film is negligible. Similar to Beer's law, the off-state transmittance of the PDLC film can be expressed as [10]

$$T = I/I_0 = \exp(-\alpha d), \quad (1)$$

where  $I$  is the transmitted light intensity,  $\alpha$  is the scattering coefficient and  $d$  is the sample thickness

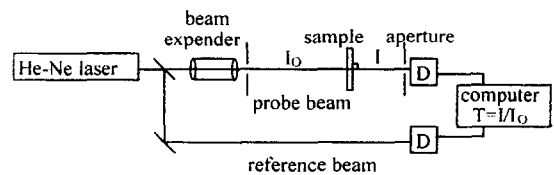


Fig. 3. Setup for transmittance measurement. D: photodetector.

of the film. The scattering coefficient  $\alpha$  is a function of the materials' properties, the NLC droplet size and the distribution of droplet axes. If two PDLC films are fabricated using the same materials and under the same conditions except alignment, the NLC droplet sizes can be conceived as the same. The main factor which makes the scattering coefficients  $\alpha$  of the two films be different from each other is the alignment of droplet axes.

For a conventional PDLC film in Fig. 2a, no alignment is made. The axes are randomly and uniformly directed to the 3-dimensional,  $xyz$ , space. Let the scattering coefficient of the conventional PDLC film be  $\alpha_3$ , then  $\alpha_3$  is independent of the polarization direction of the incident light. For a twist and a plane-parallel aligned PDLC film in Fig. 2c and Fig. 2b, the axes are directed in a 2-dimensional  $xy$  space and a 1-dimensional  $y$  space, respectively. Because their scattering coefficients depend on the polarization direction of the incident light, we let their scattering coefficients be  $\alpha_{2\parallel}$ ,  $\alpha_{2\perp}$ ,  $\alpha_{1\parallel}$  and  $\alpha_{1\perp}$ . Here, the subscript 2 represents a 2-dimensional twist; and 1 represents a 1-dimensional plane-parallel aligned PDLC film. The subscript  $\parallel$  is for the case that the polarization direction of the incident light is parallel to the  $y$ -axis; and  $\perp$  is for the case that the polarization direction is normal to the  $y$ -axis.

The refractive index of a NLC droplet is about  $n_e$  in the case that the polarization direction of the incident light is parallel to the droplet axis, and about  $n_o$  if the polarization direction is perpendicular to the droplet axis. The scattering effect comes from the refractive index mismatch between  $n_e$  and  $n_p \approx n_o$ . Because  $n_e > n_o \approx n_p$  for NLC materials, the scattering coefficients  $\alpha_{1\parallel}$  is larger than  $\alpha_{1\perp}$ . Considering that the droplets' axes are distributed in a 3- or 1-dimensional space, a simple relation for the scattering coefficients,  $\alpha_3$ ,  $\alpha_{1\parallel}$  and  $\alpha_{1\perp}$ , can be found,

$$\alpha_3 \approx (\alpha_{1\parallel} + 2\alpha_{1\perp})/3, \tag{2}$$

then

$$\alpha_{1\parallel} > \alpha_3 > \alpha_{1\perp}. \tag{3}$$

Similarly, relations for the scattering coefficients  $\alpha_{2\parallel}$ ,  $\alpha_{2\perp}$ ,  $\alpha_{1\parallel}$  and  $\alpha_{1\perp}$  in the following two extreme cases can be obtained.

Case I: when the NLC droplets axes are twisted

aligned so much, or completely randomly lie in the  $xy$ -plane, then

$$\alpha_{2\parallel} \approx \alpha_{2\perp} \approx (\alpha_{1\parallel} + \alpha_{1\perp})/2. \tag{4}$$

If  $n_e > n_o$ , then from Eqs. (2), (3) and (4), one can obtain the following relation,

$$\alpha_{1\parallel} > \alpha_{2\parallel} \approx \alpha_{2\perp} > \alpha_3 > \alpha_{1\perp}. \tag{5}$$

Case II: when the NLC droplet axes are not twisted aligned so much so that accompanied to the scattering effect the optical adiabatic effect occurs for the direct transmitted light, then

$$\alpha_{2\parallel} \geq \alpha_{2\parallel} > \alpha_3, \quad \alpha_3 > \alpha_{2\perp} \geq \alpha_{1\perp}. \tag{6}$$

In practice, a twisted aligned PDLC film should be between the conditions of Case I and Case II.

When the incident light is natural light, the off-state transmittances of the PDLC films can be expressed as

$$\begin{aligned} T &= T_{\parallel} + T_{\perp} \\ &= T_1 = \frac{1}{2}\exp(-\alpha_{1\parallel}d) + \frac{1}{2}\exp(-\alpha_{1\perp}d), \end{aligned}$$

for plane-parallel aligned film,

$$T = T_2 = \frac{1}{2}\exp(-\alpha_{2\parallel}d) + \frac{1}{2}\exp(-\alpha_{2\perp}d),$$

for twisted aligned film,

$$T = T_3 = \exp(-\alpha_3d),$$

for conventional film,

$$\tag{7}$$

where  $T_{\parallel}$  and  $T_{\perp}$  represent the transmittance of the incident light with its polarization direction parallel and perpendicular to the aligning direction of the first substrate, respectively.

From Eqs. (2), (4) and (7), and in case that the scattering coefficient  $\alpha_{1\perp} \rightarrow 0$ , one can obtain the following result,

$$T = T_{\parallel} + T_{\perp} = T_1 \approx \frac{1}{2}[\exp(-\alpha_{1\parallel}d) + 1],$$

for plane-parallel aligned film,

$$T = T_2 \approx \exp(-\alpha_{1\parallel}d/2),$$

for twisted aligned film,

$$T = T_3 \approx \exp(-\alpha_{1\parallel}d/3),$$

for conventional film.

$$\tag{8}$$

Because the fabricating conditions are all the same except the alignment, the on-state transmittances of

these PDLC films have the same value. Therefore, the PDLC film in Case I, where the NLC droplets axes are twisted aligned or randomly lie in the  $xy$ -plane, has the best contrast ratio.

However, when the incident light is linearly polarized light with its polarization direction parallel to  $y$ -axis in Fig. 2, the off-state transmittances of the PDLC films can be expressed as

$$T = T_{\parallel} + T_{\perp} = T_1 \approx \exp(-\alpha_{\parallel} d),$$

for plane-parallel aligned film,

$$T = T_2 \approx \exp(-\alpha_{\parallel} d/2),$$

for twisted aligned film,

$$T = T_3 \approx \exp(-\alpha_{\parallel} d/3),$$

for conventional film. (9)

Eq. (9) explicitly shows that the plane-parallel aligned film has the lowest off-state transmittance so it has the best contrast ratio.

Eqs. (8) and (9) are the extreme cases under the assumption  $\alpha_{\perp} \rightarrow 0$ , however, in practice  $\alpha_{\perp}$  is not small enough to be negligible.

### 3. Experiment

In the experiment, two room temperature NLCs are used: E7 and ZLI-2806 (from Merck). The main NLC is E7, a nematic mixture with clearing point at  $T_{NI} = 91^{\circ}\text{C}$ . The second one, ZLI-2806, is the host NLC for diluting a guest–host NLC, ZLI-4821/2 (from Merck), which has been reported in our previous paper [10].

The principal polymer matrix used is Norland UV-curable optical adhesive 65 (abbreviated NOA65)<sup>1</sup> the use of which has been previously discussed [10–12]. The four primary constituents of this thiol-ene system are trimethylolpropane diallyl ether, trimethylolpropane tris thiol, isophorone diisocyanate ester, and benzophenone photoinitiator.

The PDLC material is made by mixing the NLCs and the prepolymer NOA65 according to the following weight ratios: ZLI-2806(6%) + E7(54%) + NOA65(40%). With these weight ratios, the NLC

molecules can still be aligned by rubbing the substrates. In the experiment, the mixture containing the desired proportions of NLC and polymer precursors is first prepared and spread between two glass substrates which have previously been coated with a thin transparent layer of conducting indium tin oxide (ITO). The film thickness is controlled by a  $20 \pm 1$   $\mu\text{m}$  mylar spacer. To eliminate errors which come from the thickness or other fabrication conditions in the experiment, we always fabricate sample films with two kinds of alignments on the same substrates. Only half of each glass substrate is rubbed for a plane-parallel or a  $90^{\circ}$  twisted alignment, and the other half is not rubbed for making a conventional PDLC film. To make sure that the sample is uniformly aligned, we examine the sample with a cross polarizer microscope. Then the sample film is exposed to a uniform UV radiation to cure the polymer. The UV light source (Oriel Co., model 66011) is filtered by a band pass filter at wavelength  $\lambda = 360$  nm to 380 nm, and the peak value is at  $\lambda = 365$  nm. The curing intensity is fixed at  $50 \pm 0.1$   $\mu\text{W}$ , and is detected by a UV power detector arranged behind the film. The film is cured for sufficient time till the precursors polymerized enough, which is also detected by the UV power detector. When the precursors polymerized enough, the transmittance of the UV light does no longer change. During the UV curing, the sample film is put in a chamber of which the temperature is controlled at  $25 \pm 0.5^{\circ}\text{C}$ . We then examine the film using the cross polarizer microscope. The size of liquid crystal droplets in the film is uniform.

Let sample 1 be the first kind of sample, half of which is a conventional PDLC film, and the other half is a plane-parallel aligned PDLC film; and let sample 2 be the second kind of sample, half of which is a conventional PDLC film, and the other half is a  $90^{\circ}$  twisted aligned PDLC film. Their electro-optical properties are measured by the setup shown in Fig. 3. The light source in Fig. 3 is a He–Ne laser. After passing through the beam expander and the first aperture, the probe beam with spatially uniformly distributed intensity is incident on the sample film and scattered by the film. The scattered light is detected by a detector which is set just behind the second aperture. The aperture with diameter 5 mm is set 1 m apart from the sample such that the half cone

<sup>1</sup> Norland Products, Inc., P.O. Box 145, North Brunswick, NJ 08902, USA.

angle, the direct transmission of the light can be calculated by Beer's law.

#### 4. Results and discussion

Firstly, we let the light polarization direction be parallel to the alignment direction of the first substrate of the tested sample. Fig. 4 shows typical electro-optical properties of sample 1 which contains a conventional PDLC film and a plane-parallel PDLC film. From the figure, we see that the maximum transmittance of the two PDLC films is almost the same. Before reaching the maximum value, the transmittance of the plane-parallel PDLC film is always lower than the transmittance of the conventional PDLC film. That means the scattering efficiency of the plane-parallel aligned PDLC film is higher than the conventional PDLC film if the light polarization direction is parallel to the alignment direction. The off-state transmittances of the conventional and the plane-parallel PDLC films are measured as 0.0374 and 0.0279, respectively. By Eq. (1),  $\alpha = -\ln T/d$ , and because of the two PDLC films having the same thickness  $d$ , the value

$$\frac{\alpha_{1\parallel}}{\alpha_3} = \frac{\ln 0.0279}{\ln 0.0374} = 1.089. \quad (10)$$

Fig. 5 shows typical electro-optical properties of sample 2 which contains a conventional PDLC film and a 90° twisted PDLC film. Similar to the above

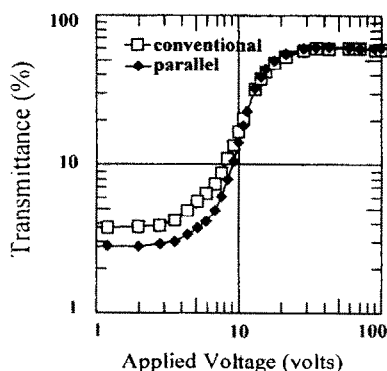


Fig. 4. The electro-optical properties of sample 1. The light polarization direction is parallel to the alignment direction of the first substrate of the tested sample. Symbols  $\square$  and  $\blacklozenge$  represent conventional and plane-parallel PDLC film, respectively.

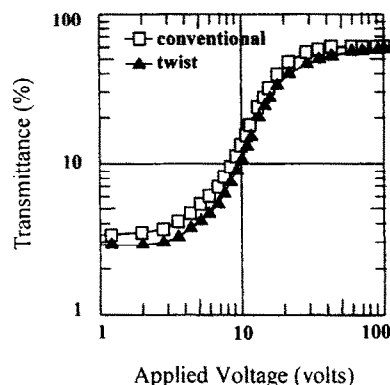


Fig. 5. The electro-optical properties of sample 2. The light polarization direction is parallel to the alignment direction of the first substrate of the tested sample. Symbols  $\blacktriangle$  and  $\square$  represent conventional and 90° twisted PDLC film, respectively.

case, before reaching the maximum value, the transmittance of the 90° twisted PDLC film is always lower than the transmittance of the conventional PDLC film. So the scattering efficiency of the plane-parallel aligned PDLC film is higher than the conventional PDLC film if the light polarization direction is parallel to the alignment direction. The off-state scattering coefficients of the conventional and the twisted PDLC films are measured as 0.0331 and 0.0284, respectively. Then,

$$\frac{\alpha_{2\parallel}}{\alpha_3} = \frac{\ln 0.0284}{\ln 0.0331} = 1.045. \quad (11)$$

From Eqs. (10) and (11), we obtain the result,  $\alpha_{1\parallel} > \alpha_{2\parallel} > \alpha_3$ , which is the same as the result predicted by theory in Eqs. (5) and (6).

We also measure the electro-optical properties of sample 1 and sample 2 with the light polarization direction perpendicular to the alignment direction of the first substrate of the tested sample. The values for the off-state scattering coefficients are listed in Table 1.

Table 1

Sample 1	Sample 2
$\frac{\alpha_{1\parallel}}{\alpha_3} = 1.089$	$\frac{\alpha_{2\parallel}}{\alpha_3} = 1.045$
$\frac{\alpha_{1\perp}}{\alpha_3} = 0.962$	$\frac{\alpha_{2\perp}}{\alpha_3} = 0.965$

The results in Table 1 show that  $\alpha_{1\parallel}/3\alpha_3 + 2\alpha_{1\perp}/3\alpha_3 = 1.004 \approx 1$ , in agreement with Eq. (2). Also from Table 1, we obtain the result  $\alpha_3 > \alpha_{2\perp} \geq \alpha_{1\perp}$ , which is the relation described by Eq. (6). That means, accompanied to the scattering effect, the optical adiabatic effect occurs in the 90° twisted PDLC films.

From the experiment and theory, we believe that a highly twisted aligned PDLC film can have higher contrast ratio than a conventional PDLC. However, such a PDLC film shows hysteresis of the memory effect [13]. There is another way to have all the NLC droplets' axes lying in a plane parallel to the substrates. That is to use dual frequency NLC. One can apply a low or high frequency ac electric field for alignment when curing the PDLC film, and drive the film at a high or low frequency.

## 5. Conclusion

To increase the contrast ratio of a PDLC film, we study the alignment for PDLC films. We propose a theory based on Beer's law for describing the off-state transmittance of each kind of alignments. The theory suggests that the PDLC film with all of its NLC droplets' axes lying in a plane parallel to the substrates can have higher contrast ratio than a conventional PDLC film. Such PDLC films can be made by using a highly twisted alignment, or by using the dual frequency NLCs and aligning the droplets' axes by the electric field. The experimental results fall

between the two extrem cases described in our theory.

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